

INTRACRYSTALLINE DIFFUSION IN CLINOPTILOLITE:
IMPLICATIONS FOR RADIONUCLIDE ISOLATION, Sarah K. Roberts, Brian E. Viani, and Douglas Phinney, Lawrence Livermore National Laboratory, Livermore, CA.

Experiments have been performed to measure the rate of exchange diffusion in the zeolite clinoptilolite (CL) for elements important to radionuclide isolation at Yucca Mountain, NV. Single crystals of CL were rendered nominally homoionic (shown by electron microprobe analyses) with respect to Ca, Na, and K by exchange with solutions of CaCl_2 , NaCl, and KCl. The crystals were mounted to expose a single crystal face, and were immersed in solutions of Cs+cation and Sr+cation (cations = Ca, Na, K) at ambient (30°C) and elevated (90°C) temperatures. The modeling code, EQ3/6, was used to select solution compositions that would maintain a constant concentration boundary condition at the crystal surface and that would result in the inward diffusing cation occupying approximately 1% of the exchange sites at the crystal/liquid boundary. After varying periods of contact, the crystals were removed from solution and concentration profiles of the inward diffusing cation normal to the exposed face were measured by Secondary Ion Mass Spectrometry. A binary interdiffusion model, in which the effective interdiffusion coefficient is dependent on the composition of the exchange sites, was used to model the ambient temperature diffusion of Cs and Sr into Ca - CL crystals. A fit of this model to preliminary data indicated that at 30°C Cs and Sr tracer diffusion coefficients are on the order of 10^{-13} and $10^{-18} \text{ cm}^2\text{s}^{-1}$, respectively. These results indicate that under certain conditions, exchange equilibrium cannot be assumed when modeling transport of Cs and Sr. Similar analyses for diffusion of Cs and Sr into Na and K - CL will also be presented.

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